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LLNL-JRNL-650120

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February 18, 2014

Applied Physics B

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Applied Physics B manuscript No.
(will be inserted by the editor)

Measurement of femtosecond atomic lifetimes using ion traps

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Received: date / Accepted: date

Abstract Two types of experiment are described that both employ an electron beam ion trap for the production of highly charged ion species with the aim of then measuring atomic level lifetimes in the femtosecond range. In one experiment (done by Beiersdorfer et al. some time ago) the lifetime measurement depends on the associated line broadening. In a recent string of experiments at LCLS Stanford, the HI-LIGHT collaboration employed pump-probe excitation using the FEL as a short-pulse X-ray laser.

Key words Oscillator strengths, lifetimes, transition moments Ion trapping

1 Introduction

Ion traps are regularly employed to store ions for extended periods, so that they can be interrogated ever so often or remain unperturbed for a longish time. The longest level lifetime measured in a singly charged rare earth ion appears to have been on the order of ten years [1], interrogating a stored ion by laser for the population of a specific high angular momentum level. The bulk of atomic lifetime measurements on trapped ions has dealt with level lifetimes in the range of about one minute down to hundreds of nanoseconds [2]. The latter experiment operated by suddenly stopping the excitation by an energetic electron beam and measuring the decay of the fluorescence signal by electronic timing. (Along the way, quite a number of such measurements have employed a radiofrequency or Paul trap, usually for lifetimes in the millisecond range.) Electronic timing on a single-nanosecond scale after excitation by laser light has been a standard technique used in many

lifetime measurements on neutral atoms. Fast-ion beam techniques (foil, gas, or laser excitation of an ion beam at kinetic energies of keV through GeV) replaced the electronic timing by spatially resolved observation as the ions travel on after leaving the interaction zone (for a typical example, see [3]). The shortest lifetimes measured with foil-excitation and spatially resolved observation are on the order of one picosecond, requiring foil displacements and control over the observation geometry on the order of a few micrometers. Geometry was exploited differently when measuring the (few dozen femtosecond-) lifetime of K-vacancies of ions travelling in solids by using exciter foils of different thickness [4]. Inside the solid matter, the interplay of collisional ionization, excitation, and electron capture shifts the ionization balance in correlation with the electron core level lifetimes. Autoionizing states of various ions feature lifetimes well below 1 ps. Beam-foil spectroscopy has tried to obtain valid data on those sub-ps lifetimes by using the foil as an interaction medium, but then evaluating line broadening as an indicator of a very short atomic lifetime, preferentially of the lower level of a transition so that the fluorescence yield for optical observation of the upper level decay would be high [5]. Overviews over lifetime measurement techniques using fast ion beams (among others [6,7]) and ion traps (among others, [8–13]) are available and give numerous further references, as well as a discussion and examples on how to reach for accuracy in atomic lifetime measurements of multi-electron heavy ions and what has been achieved.

Evidently, the first femtosecond atomic lifetimes have been measured already decades ago, so why is there any interest in returning to the topic, and why would one do it using an ion trap that seems designed to deal with much longer time scales than asked for in the measurement of extremely short atomic level lifetimes? Firstly, the early optical experiments suffered from line broadening due to the excitation process, rendering the measurements of atomic line broadening much less accurate (typically not much better than the order of magnitude) than one had hoped for. Since then we have learned to produce ions of any charge state of any element, be it by fast-ion techniques or inside an electron beam ion trap. Much better control over the excitation conditions is available now, especially in electron beam ion traps (EBIT) [14–19]. Secondly, at a time when most atomic lifetime research dealt with lifetimes in the range from a few picoseconds to nano- or microseconds, femtosecond decays seemed exotic, and a rough estimate backed by experiment was good enough. However, highly charged ions feature mostly very short atomic level lifetimes, and the steep scaling of most transition rates with the nuclear charge Z brings many key levels of heavier ions into the femtosecond range, for atomic systems that play important roles in astrophysics or terrestrial fusion experiments. We now want accurate data also in this range that will help us understand peculiarities in hot terrestrial and astrophysical plasmas, from the solar corona to the accretion disk around a black hole in the center of a galaxy.

With highly charged ions, so far two series of experiments have emerged that both depend on an electron beam ion trap for the production of the de-

sired ion species so that then atomic lifetimes in the femtosecond range can be measured. In one experiment (passive observation) the measurement depends on the associated line broadening, in the other (active interrogation), a short-pulse X-ray laser is used in pump-probe experiments.

2 Experiments using line broadening

An electron beam ion trap confines charged particles by a combination of a Penning ion trap (ring electrodes on different electrical potentials aligned with a strong magnetic field) and a high-density electron beam (further ionizing trapped particles, attracting positively charged particles, partly compensating the repulsive space charge of the positive ions). The trap depth is largely given by the ion charge and the potential drop between the electrodes (drift tubes). Evidently more highly charged ions experience a deeper potential well than others. This leads to the preferential trapping of heavier elements that can be more easily ionized, up to the limit prescribed by the ionization potentials of successive ionization stages in relation to the electron beam energy. Because of their particularly high ionization potentials it is possible to not only have ions with noble-gas like electron configurations as the highest charge state present, but also to reach a very high percentage of these ions in the charge state distribution.

Although the trapped ion cloud overall is practically at rest, the ions in the cloud move according to their energy; the cloud has a temperature, and high-resolution spectroscopic observations suffer from the Doppler broadening of the spectral lines. However, the hottest ions may overcome the potential barriers and escape, reducing the temperature of the remaining ions in the process. Mixing in light elements helps with this evaporative cooling of the ion cloud. A further temperature reduction is achieved by lowering the drift tube potential differences to zero or even to negative values [20, 21].

Following this recipe, the Doppler width in X-ray observations of (Ne-like) Cs^{45+} ions was reduced substantially and made significantly smaller than the instrumental line width and the natural line width of the $3d_{5/2}$ level. This process can be followed by simultaneously observing a nearby spectral line that originates from the decay of the much longer lived 3s level which does not feature notable lifetime broadening under the circumstances. A resolving power of $E/\Delta E = 2\times\ 10^4$ was achieved. A comparison of the line widths yields the contribution of lifetime broadening and after inversion a level lifetime of barely 2 fs, with an uncertainty of -3+13%, which was in the ballpark of the scatter of several predictions of the time [22]. The asymmetry of the error bar results from the systematics of the data evaluation, extracting a change in line width, and the wanted entity being derived from the increase to which it had added in quadrature.

Theory may be expected to be more reliable for He-like ions (two electrons) than for Ne-like ions (10 electrons), while the experimental technique

should be equally applicable. However, a test experiment on the 1s2p levels of Fe^{24+}, an ion species that figures prominently in plasma physics research and in astrophysical sources, was interpreted [23] to yield a result (obtained via two different paths) at variance with the most trusted calculations [24]. Meanwhile a re-analysis of the experimental material has identified a data processing problem as the source of the discrepancy; the data now point to a result that fully agrees with the theory results of just under 2.2 fs [25].

3 First ion trap and X-ray laser experiments at LCLS

The HI-LIGHT collaboration (EBIT groups at MPIK at Heidelberg, Germany, and at Lawrence Livermore National Laboratory, CA, USA, plus associates) has set out on a quest to provide a target of highly charged ions (in an ion trap) for the world's most brilliant X-ray laser (LCLS), and to study key physics problems that have not been accessible in any other way. A transportable EBIT previously used at the Free-Electron Laser at Hamburg (FLASH) and thus dubbed the FLASH-EBIT (and also used at synchrotron light sources such as PETRA and BESSY II) was set up at LCLS and complemented by equipment from Livermore, especially a cryogenic microcalorimeter for versatile X-ray measurements. FLASH-EBIT provides ions in a pencil-shaped horizontally oriented cloud a few centimeters long and about half a millimeter wide, resulting in a target of areal density $10^{10}/cm^2$. The ion trap was surrounded by various photon detectors (Germanium detectors, a microcalorimeter, an XUV spectrometer with an MCP detector, etc.) and an ion extraction system feeding a particle detector. LCLS delivered narrowband photon pulses with a pulse length between some 200 fs and several picoseconds at a rate of 120 Hz; the energy per pulse amounted to about 3 mJ, with X-ray energies adjusted in the range from 600 to 900 eV. The X-ray light was focused to a waist less than 100 μm wide and traveled along the axis of the ion cloud. The width of the target ion cloud was known from estimates based on optical observations and was now verified by monitoring the X-ray fluorescence while sweeping the X-ray pencil beam across the target.

Sub-microsecond time resolution was essential to improve the signal-to-noise ratio. The demonstration experiment on the resonant photo-excitation of astrophysically important of two $2s^2\ 2p^5\ 3d$ levels, $^1P^o_1$ (at 825.791 eV) and $^3D^o_1$ (at 812.425 eV), of Fe^{16+} ions has already been successful [26]. It indicated that a persistent discrepancy between theory and astrophysical observations for the relative intensities of a prominent pair of lines in the soft-x-ray range was not to blame on the astrophysical environment and imperfect collisional-radiative modeling, because a clean laboratory experiment corroborated the astrophysical observations. Transition rates of electric dipole (E1) transitions depend on details of the (nonorthogonal) radial wave functions and therefore test atomic structure details beyond the energy levels. In this case, the upper level (singlet-triplet) mixing is

known from accurate spectroscopy and atomic structure theory, yet even extremely large computational basis sets and involved calculations have not been able to reproduce the experimental findings of a conceptually simple experiment that uses resonant photoexcitation. Shortcomings in the atomic wave functions used in the atomic structure calculations now appear as the likely cause. However, in spite of renewed efforts, a solution to this atomic structure calculation challenge has not yet been found.

More experiments have been undertaken by the HI-LIGHT collaboration (data evaluation is under way), and others are planned. Here only experiments addressing atomic lifetimes in the femtosecond range are to be discussed. As indicated above, lifetime measurements may be able to shed more light on the discrepancy between measurement and computation of the aforementioned prominent soft-x-ray line intensity ratio that involves two excited levels with lifetimes in the femtosecond range.

4 Pump-probe X-ray laser experiments at LCLS

Pump-probe experiments have been very successful in the study of chemical reactions or in the investigation of details of how wave functions evolve after perturbations, and the value of the measurement technique has been acknowledged by the Nobel Prize for Ahmed Zewail. With highly charged ions the challenge lies in the fact that the first excitation step requires a high photon flux at X-ray energies, to be followed by another intense light pulse in either the same wavelength range or any other, depending on the structure of the ion studied. The Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Center (SLAC, at Menlo Park, CA, USA) operates a pump-probe set-up in one of their experiment hutches which allows the user to heat a solid target by an intense IR laser and then interrogate it by an X-ray flash from the 1 km long free-electron X-ray laser. By changing the time difference, it should be possible to also first excite by X-rays and then manipulate the excited material by the IR laser. However, to atomic physicists it is more appealing to study individual atoms and ions and their interaction with narrowband light. This is the topic of a test measurement already tried and of the HI-LIGHT plans developed since.

The aforementioned striking outcome of the demonstration experiment [26] calls for further tests of the same atomic system, for example measuring the decay rates of the two excited levels involved, or their inverse, the level lifetimes, which are expected near 44 and 166 fs, respectively [27,28].

Pump-probe experiments in atoms try to use laser light that is tuned to each of the transitions of interest (two-colour pump-probe). This is not yet feasible at LCLS (but the laboratory is working towards such capabilities). For the first round of experiments a double pulse technique was used that used the X-ray light from LCLS for both light pulses (single colour pump-probe). The atomic physics scheme is shown in figure 1. The electron beam ion trap is set to provide Fe^{16+} as the highest charge state present. The

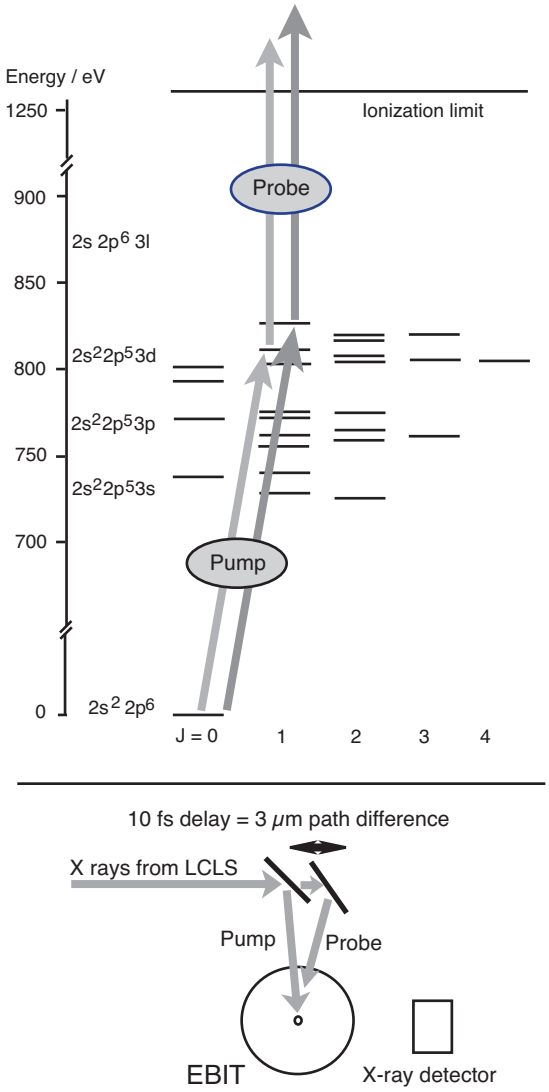

Fig. 1 Schematic of a pump-probe experiment on Ne-like iron ions (Fe^{16+}, simplified level scheme in the upper part), assuming a beam splitter for X-rays (lower part). The two $2s^2\ 2p^5\ 3d$ $J = 1$ levels of primary interest have predicted lifetimes of 44 fs and 166 fs, respectively.

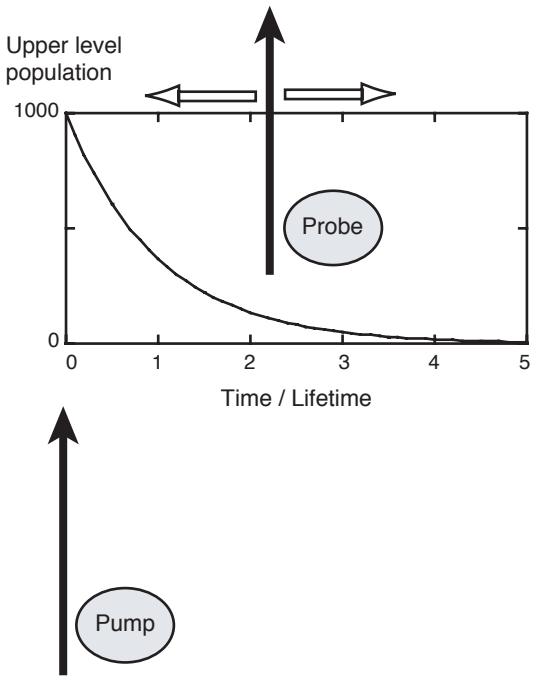

Fig. 2 Schematic of a pump-probe experiment for atomic lifetime measurements. The first pulse excites a sample the emission signal of which then decays exponentially. A second light pulse probes the remaining excited level population and, by varying the time delay after the first pulse, maps out the exponential curve from which the decay rate can be derived.

impinging X-ray photons resonantly excite the level of interest (confirmed by fluorescence spectra), but cannot ionize the target ion directly. The second light pulse may excite further ions from the ground state or further ionize ions that are still excited. Then (and only then) Fe^{17+} ions are produced which (ever so often, say, every few minutes) may be extracted (slowly, over several seconds, for maximum efficiency and clean spectra) from the trap and detected. A series of measurements at different time differences can then map out the population history of the excited level in Fe^{16+}, from which decay curve the level lifetime may be determined (see schematic in figure 2). The first such measurement has been attempted in 2012.

The measurement principle evidently is simple. Even if during the first light pulse a second photon would hit the excited ion and ionize it, this would only add a constant background to the data. This background can

be estimated from experiments with single pulses of constant power and of varying duration. The signal should grow with the pulse power until the excitation saturates (with half of the ions in the excited state). Although some experts advised that this would happen at the high intensity of the LCLS photon beam, it did not happen in this experiment which, by the way, did not aim for a particular tight focus, since the ion target is much wider than the few-μm focus LCLS can achieve. Hence it would be easy to increase the X-ray flux density, but the flux would then pass through fewer target ions. To achieve a higher signal, the total X-ray flux has to be increased, which is an ongoing pursuit at LCLS.

The actual femtosecond timing element in this experiment is not in the detection, but in the excitation. In many double-pulse experiments the two light pulses are produced by using a beam splitter with a displaceable mirror segment, employing a longer path for part of the beam to make it arrive later on target. (The speed of light is 300 μm/ps.) A first such split-mirror system has meanwhile been activated at LCLS, but so far none is available for the X-ray range needed here. Instead a scheme has been tried that had been suggested by the LCLS accelerator team and is based on a method used previously for the production of single-femtosecond pulses [29] (and that borrows from ion accelerator techniques for producing short particle bunches for timing purposes in nuclear physics, especially for neutron reactions, developed in the 1950s). The electron beam of the accelerator consists of many short bunches matched to the high-frequency acceleration scheme. In order to correct for dispersion errors, there are short sections between the long accelerator sections in which a set of four magnets first deflects the electron beam and then brings it back onto its trajectory. This sideways bend-and-bend-back arrangement is called a *chicane*, and it is being used also to measure and constrain the physical length of the electron pulses before they enter the free electron laser (FEL). During the deflection, as a consequence of some momentum spread within the bunch, a given electron bunch is no longer aligned with the beam trajectory. The slanted electron bunch is directed through a thin foil (the "spoiler") with a small opening that lets pass only part of the electron bunch unharmed (and spoils the narrow momentum distribution of the rest that then will be lost from the beam). When the surviving electron bunch is used in the FEL to produce an X-ray flash, the shorter bunch results in a shorter X-ray flash. Of course, fewer electrons in a shorter bunch also imply much less gain in the lasing process.

For our experiment the single opening was replaced by two slots in a foil (see figure 3). If the electron bunch reaches across both slots in the "slotted spoiler", the slots provide two openings that let parts of the bunch pass through. The slots are not parallel to each other. By displacing the obstacle foil sideways, different distances between the openings can be arranged which translate into different time separations (some 80 to 300 fs) of the two short X-ray pulses produced farther down the line. Although the geometrical idea is straightforward, the device needs to be calibrated, for example

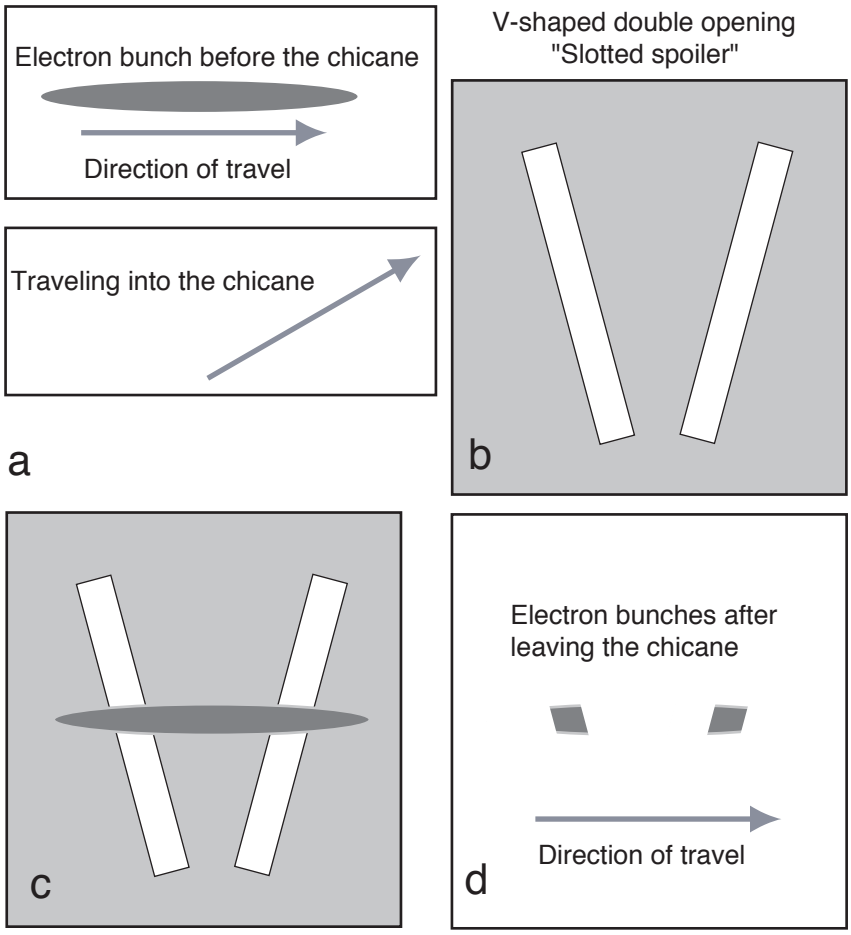

Fig. 3 Slotted spoiler production scheme for two short fs-pulses of adjustable time difference from a single electron bunch (a) arriving at the chicane of a free-electron laser. For the process, the electron bunch is being rotated in space (b) so that it travels towards the spoiler foil (plane of paper). The time difference is realized by a (vertical) displacement of the foil with the two slots (c). Those parts of the electron bunch that interact with the foil material will be lost, and only the two short sections that have passed through the cut-outs are rotated back and continue their flight (d) to the FEL.

by studying an atomic lifetime of the same magnitude in a simpler atomic system for which theory can be trusted to be accurate. Such references are available in the $n = 2$ resonance levels of the one- and two-electron ions of lighter ions, say of fluorine, neon, and magnesium, which have excitation energies and level lifetimes compatible with LCLS operation parameters. For example, the lifetime of the 1s2p $^1P^o_1$ level in He-like Ne^{8+} ions is predicted to be 113 fs [24] and thus commensurate with the 44 and 166 fs level lifetimes predicted for the two $J = 1$ levels of interest in Fe^{16+}.

For a first-of-its-kind experiment in this wavelength range, there are many unknowns to the functionality of the concept. Rather conservative estimates projected a signal rate of one detected Fe^{17+} ion per 100 LCLS cycles at the full X-ray beam power or a sizeable fraction thereof, which would yield 10^5 counts per 24-h day and result in less than 1% statistical uncertainty of the desired atomic level lifetime, in the absence of background and systematic errors (which would need to be explored). As it happened, the double light pulse production seemed to work, but the aperture slots each transmitted only some ten percent of the total electron bunch, and thus each of the X-ray flashes had an energy of only a small fraction of the full-length light pulses. This power level turned out too low for the intended measurement. (A lightning-induced blackout of the whole laboratory which required several days of recovery effort and worsened the run conditions afterwards did not help either.) However, the non-success of the pump-probe attempt using pulses made by the slotted spoiler technique does not invalidate the concept. The experiment seems worth trying again when a more efficient means of production of X-ray pulse pairs with femtosecond spacing becomes available.

5 Other schemes

Meanwhile other schemes are being investigated theoretically by the HI-LIGHT collaboration that combine atomic structure studies with atomic level lifetime measurements. One problem so far is the low ion yield mentioned; it would seem advantageous to increase the signal rate substantially. One approach would be a pump-probe experiment in which the probe laser light is in the IR, visible, or UV range and would be in resonance with an atomic transition and thus achieve large production cross sections. The probe laser would transfer population to a level that in turn decays in the X-ray range and can be detected with a higher yield than the detection of specific ions achieves. One can also search for a second X-ray pulse of a wavelength that differs from the first and connects to an excited level in the next higher charge state ion, taking advantage of resonance effects in photoionization. LCLS is already trying out two-colour operation, expanding their toolkit towards the variety long since established in visible-laser spectroscopy, but now applied to X-rays and the femtosecond time range. LCLS so far is unique (and evolving) in this domain, and so is the electron

beam ion trap as a producer (and possible target) of highly charged ions. Wolfgang Paul surely would enjoy this combination.

Acknowledgement

The author appreciates thankfully the hospitality and support experienced at the Livermore EBIT laboratory and the work experience in the HI-LIGHT collaboration. Part of this work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Support by the German Research Association (DFG) is gratefully acknowledged (Tr171-19).

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